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Capture cross section of 14 MeV neutrons

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Radiative capture cross sections for some nuclei round about mass number 100 have been measured for 14·8 MeV neutrons by the activation technique Sources of systematic errors and the steps taken to control them have been discussed. The results have been compared with those given by Perkin *et al* and Csikai *et al*. Data obtained are very much different from those predicted by the compound nucleus theory.

INTRODUCTION

Neutron capture by the nucleus is a complicated process. To understand the reaction mechanism, experimental investigations were mostly performed by using thermal neutrons, since they have the larger cross sections, and by studying the gross structure of the capture γ-ray spectra. Nevertheless, total cross sections of (n, γ) reactions of higher energy neutrons, especially 14 MeV neutrons have also been reported by a few workers (Perkin *et al* 1958, Cvelbar *et al* 1966 Csikai *et al* 1967) Comparisons of such data have been made with those obtained from calculations according to simple theoretical reaction models. Experimental cross section values of 14 MeV neutrons, captured by nuclei of mass number higher than 50, are generally, a few orders of magnitude larger than those obtained from calculations according to statistical model (Lane & Lynn 1959). A so called 'semi-direct' capture model (Brown 1964, Clement *et al* 1965) raises the theoretical values in some cases by a few factors of ten, near 14 MeV But these values also fall shorter by about a factor of ten than the reported experimental values (Csikai *et al* 1967). Moreover, not many experimental values exist for neutrons over 10 MeV of incident energy for any meaningful comparison. Therefore, mainly to supply a set of reasonably accurate experimental values, the following work was undertaken.

The method followed for this work has been the conventional activation measurements. Following such a method, results in the past were presented by Perkin *et al* (1958) and later by Cvelbar *et al* (1966) and Csikai *et al* (1967). Here the values of total cross sections of (n, γ) reactions of 14 MeV neutrons are presented for the following nuclei above mass number 50, *viz.* ^{71}Ga, ^{75}As, ^{127}I, ^{138}Ba and ^{141}Pr.

EXPERIMENTAL METHOD

The $\sigma(n, \gamma)_{total}$ was determined from relative measurements of saturation β^- or β^+ activities developed after long neutron-irradiation of sample elements

Generally from a composite decay of 'daughter' element, estimate could be made of the saturation activity of the sought-for isotope developed by (n, γ) reaction. As the neutron source the $(T-D)$ generator of Bose Institute was used.

Systematic Errors : Naturally, when the cross section values are of the order of magnitude of only fractions, *or* at best a couple of units of millibarns (as predicted by model calculations or previously obtained results, by other workers) one must have to take extreme caution to minimize and/or eliminate every possible error involved. In the present case, errors were held to be composed of the following ones :

(a) Error due to indefiniteness of the incident neutron energy.

(b) Error involved in relative β—counting due to mainly the self-interaction of the β-rays.

(c) Error due to masking caused by activities present from reactions other than (n, γ),

(d) Error due to variation of neutron-flux, which is by far the most serious error.

Procedures followed to eliminate or to minimise the above errors are briefly described here.

(*i*) The energy spread of incident neutrons were determined knowing exactly the geometry, *i.e* , from the measured dimension of focal spot on the tritium target and the area and the distance of the sample, and then estimating from the reaction kinetic calculations. The H.T. source of the neutron generator was stabilized and the voltage variation thereby was restricted to within $\pm 0.1\%$ over 150 KV. This comparatively low H T. value was used for neutron generation since the spread on the sample was rather large, $= 30°$; hence to restrict the variation within a low limit. 120-150 KV were the compromise values. The large spread was to obtain larger counts, because the approach of sample to source had to be close. The thickness of the sample again was also a compromise, rather then being unduly thick. The effect of this sample thickness is considered separately in (ii). The value of incident neutron energy thus specified was 14.8 ± 0.08 MeV.

Needless to say, the system of irradiation was kept 'clean', having minimum scattering material nearby. For this reason, liquid coolant in the target was dispensed with and forced air cooling was used. This necessitated some inventiveness to prevent fly-out of (generally used) powdered samples, which were irradiated without any cover. No extra backing was used for the thin copper disc of the titanium tritium target foils.

(*ii*) *Errors in relative β-counting involved* :

In one of the earlier papers published from this laboratory (Mitra & Ghose 1966), the sources of error associated in β-counting of such thick samples, were

discussed and methods to eliminate or minimize them pointed out. However, for the sake of completeness, they are very briefly discussed here. Most of the correction factors involved in thick-sample β-counting were calculated and employed in a straightforward manner, except self-absorption and self-scattering in the sample, which contribute to the largest errors. These self interaction factors were determined by a procedure which was a variation from those adopted by Prestwood & Bayhurst (1959). Overall relative efficiency ϵ_{rel} of β-counting was obtained from a semi-empirical relation

$$\epsilon_{rel} = 1 - \exp(-b\bar{E}_\beta)$$

where $\bar{E}_\beta$ is the *average* energy of β, b is a constant which was experimentally determined for a constant geometry of source and counter during counting Although such a relation slightly deviates from general, at very low values of $\bar{E}_\beta$, the deviation was seen to be within a few percent, and the overall relative efficiency of the particular set-up of counting was determined within $\pm 3\%$ of the total for the $\bar{E}_\beta$'s encountered in all the cases of our sample activities.

(*iii*) *Masking caused by other activities* :

It is difficult, in these high mass number elements to get rid of or accurately estimate the lingering activities present due to $(n, 2n)$ reaction products, since in nearly all the cases, the $\sigma(n, 2n)$ exceeds the $\sigma(n, \gamma)$ by a factor of 10^2-10^3, in this neutron energy. However, accurate follow-up of the composite decay curves (figure 1) and reconstruction of the wanted activity from them by adopting a procedure of fitting by least square method, yielded results of desired accuracy. Parallelactivities of comparable half lives were absent in the chosen samples reactions The $(n, 2n)$ caused, in general, increased backgrounds and most of the errors in counting were due to difficulty in estimating the small activity present from large backgrounds

(*iv*) *Standards* :

The standard reactions adopted for relative measurements were $(n, 2n)$ reaction in ^{63}Cu, adopted value of cross section being 530 ± 25 mb (Mitra & Ghosh, 1966) and (n, α) reaction in ^{27}Al, the adopted value for this σ was 116 ± 8 mb The former reaction was adopted for comparatively short half lives of products and the latter for longer half lives.

(*v*) *Neutron flux variation* ·

Neutron flux was continuously monitored by a heavily biased plastic scintillator detector connected to a sensitive count rate meter and recorder. Flux could be held constant to a deviation of $\pm 5\%$.

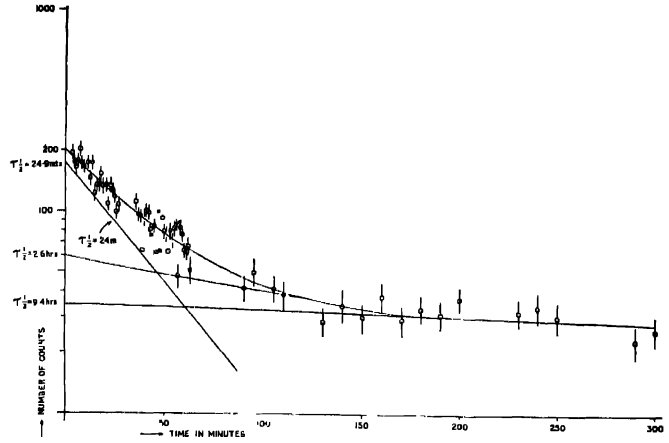

Figure 1. Analysis of the composite decay curve of ^{127}I to extract different activities.

(*vi*) *Experimental details* :

Samples were taken in powder form and the chemical form used were respective oxides. Purity of the samples were taken as guaranteed by the manufacturers. The sample holders were in the form of a circular pot of diameter 2.5 cms, in which samples could be pressed to form tablets of maximum thickness 2 mm. Holders were made from pressed graphite. Plastics were not used because of their possible effect of moderating neutron energies to lower values, although during the counting of the long-lived samples, they were often emptied into a plastic pot of same dimensions. Sample to source distance was always maintained to be 4.5 cms in the forward direction. Irradiation was given, keeping the samples inside cadmium boxes. Foils of specpure copper and aluminium as standard materials, were given exact circular diameters as sample-cakes and they were placed above and below the samples. The standard foils were each of 10mg/cm^2 thickness. A separate correction was made for difference in geometrical dispositions of the sample and target foils towards the neutron source spot.

For iodine samples, during counting, care was taken to ensure that there was no loss by sublimation or that it did not deposit itself on the counter window. For this, a special ice-box was constructed (figure 2) and iodine, after irradiation being transferred in a weighed perspex pot, was housed in the ice-box and counted. Any sublimate depositing on the counter window was checked for, simply by removing the sample and looking for background counts. It was observed that if the ice-box was used, no extra correction was needed for loss of

sample or its deposition on the window, the effects being eliminated. A dry chamber used for 'sample to counter volume' eliminated deposition of moisture on the sample.

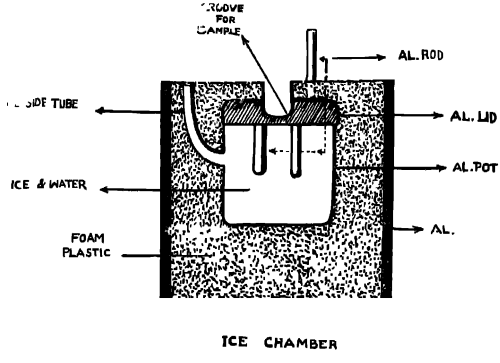

Figure 2. Diagram of ice chamber developed and used during the counting of ^{127}I.

The counter used was end window β-counter of 2.4 mg/cm^2 window thickness; sample to counter distance was 1cm.

Results

The total $\sigma(n, \gamma)$ was calculated by the usual method, from saturation activities in the sample and the standard, taking into account all the errors discussed above Ideas of expected half lives were obtained in each case from the data supplied in the table of isotopes by Hollander *et al* (1967), then all re-determined by constructing the full decay curves.

(*i*) *Irradiation of gallium* : Specpure Ga_2O_3 (white powder) was used as sample. Standard in this case was ^{27}Al, in the form of specpure foil. Sample consists mostly of two isotopes ^{69}Ga and ^{71}Ga, in the ratio 39.5 : 60.2 The life-times of generated activities were therefore expected as, 32 secs from ^{68}Cu, by (n, α); 2 2 mts from ^{71}Zn, by (n, p); 5.1 mts from ^{66}Cu by (n, α); 21.1 mts from ^{70}Ga by $(n, 2n)$; 57 mts from ^{69}Zn and 68 mts from ^{68}Ga These half lives were confirmed by actual follow up of the decay curve. None of them really competes with 14.3 hours activity obtained from ^{72}Ga. However, a large background due to ^{68}Ga and ^{70}Ga was present from which 14.3 hours activity was separated Multiple irradiation of 2 5 hours each was given for saturation of long time activity.

(*ii*) *Irradiation of arsenic* : ^{75}As is a 100% isotope. The activities generated after long irradiation were : 82 mts due to ^{75}Ge by (n, p); 14.3 hrs due to

^{72}Ga by (n, α) and 26.5 hrs due to ^{76}As by (n, γ); sample used was As_2O_3 of 99 5% purity. ^{27}Al (n, α) was the standard.

(*iii*) *Irradiation of iodine* : Iodine metal flakes, pasted in the form of cakes were used as sample. ^{127}I being a 100% isotope, only activities expected were 9.4 hrs from ^{127}Te, by (n, p); 13.3 days from ^{126}I by $(n, 2n)$; 25 mts from ^{128}I by (n, γ). From the decay curve, a new activity of 2.6 hrs was recognized and computed. Aagard *et al* (1957) reported such an activity in their study of fission of ^{126}I This was recognised as due to the isomerism of 13 days $(n, 2n)$ product. $(n, 2n)$ in ^{63}Cu was the standard.

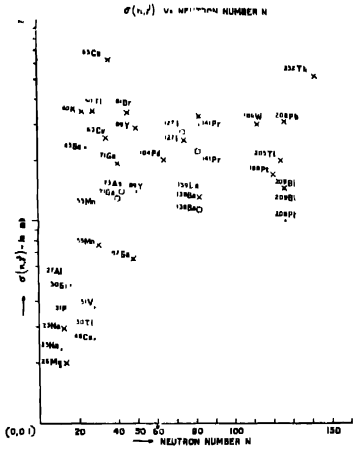

Figure 2. Variation of $\sigma(n, \gamma)$ with neutron number is shown.

(*iv*) *Irradiation of barium* : There are too many isotopes in the element barium, starting from 130 to 138 mass number barring 131 and 133; the largest percentage is that of ^{138}Ba, being 71.66. Hence the decay curve expected was really a complicated one. But in reality the 84 mts half life of ^{139}Ba was easily recognized since $(n, 2n)$ reactions of principal isotopes give all stable products. Only competing activities arose from (n, p) of ^{138}Ba *i.e.*, ^{138}Cs. Irradiation time was 168 mts. ^{63}Cu$(n, 2n)$ was used as the standard.

(*v*) *Irradiation of praseodymium* : ^{141}Pr is again a 100% stable isotope. The activities obtained were 5.4 mts from ^{140}Pr, by $(n, 2n)$ and 19.2 hrs from ^{142}Pr by (n, γ). The sample was specpure Pr_2O_3 black oxide powder. The standard was (n, α) in ^{27}Al.

TABLE 1. Capture cross sections for 14.8 MeV neutrons

	Reaction	Target	Neutron no. N	σ in (mb) $\pm$ counting error	Monitor reaction	Author	Values calculated (compound nucleus theory, Lane 1957)	Particles counted
(1)	$^{71}Ga(n, \gamma)^{72}Ga$	Ga_2O_3	40	$1.90 \pm 10\%$ $1.29 \pm 15\%$	$^{27}Al(n, \alpha)$	Perkin *et al* (1958) Present author	0.1 mb	β^-
(2)	$^{75}As(n, \gamma)^{76}As$	As_2O_3	42	$1.39 \pm 12\%$	$^{27}Al(n, \alpha)$	Present author	0.11mb	β^-
(3)	$^{127}I(n, \gamma)^{128}I$	Iodine	74	$2.50 \pm 10\%$ $2.74 \pm 7\%$	$^{63}Cu(n, 2n)$	Perkin *et al* (1958) Present author	0.14mb	β^-
(4)	$^{138}Ba(n, \gamma)^{139}Ba$	$BaCO_3$	82	$1.30 \pm 30\%$ $1.12 \pm 20\%$	$^{63}Cu(n, 2n)$	Perkin *et al* (1958) Present author	0.14mb	β^-
(5)	$^{141}Pr(n, \gamma)^{142}Pr$	Pr_2O_3	82	$3.33 \pm 10\%$ $2.19 \pm 14\%$ $3.00 \pm 10\%$	$^{27}Al(n, \alpha)$	Perkin *et al* (1958) Present author Csikai *et al* (1967)	0.14mb	β^-

The values of $\sigma(n, \gamma)$ obtained for all these isotopes are presented in table 1, along with the values obtained by other workers.

Discussion

In all the cases of irradiations, the effects of rarer types of reactions, *viz* (n, d), (n, t) etc were ignored, since their reported cross sections in the isotopes studied were less than orders of magnitude, as the daughter products are stable or their cross sections have not been reported. It is seen that there is an increasing tendency of the cross sections with increasing mass number, *i e*, with increasing number of neutrons. A plot of $\sigma(n, \gamma)$ *vs* neutron numbers (figure 3) has been done.

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